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QUASI-WHISPERING GALLERY MODES IN ZnO MICROTOWERS

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Abstract

We show that the laser spectra of ZnO films composed of particles that look like microtowers represent two bands with practically constant spectral distances between their peaks. By numerical analysis, we show that these two bands are due to whispering gallery modes (WGM), and one of these bands corresponds to the quasi-whispering gallery mode. Thus, we obtain here the first clear demonstration of the existence of quasi-WGMs, and show, in addition, that the width of the bands is caused by random spread of sizes of the ZnO particles in the lasing part.

Keywords: zink oxide, ZnO film with microtowers, laser spectra, whispering gallery modes (WGMs), quasi-WGMs, numerical analysis.

1. Introduction

The wide-band-gap (3.3 eV at room temperature) semiconductor ZnO provides a possibility for the formation of rich micro/nanostructure diversities, many of which may be suitable for lasing action. Therefore, the analysis of laser modes in these structures is of certain interest. Since the ZnO refraction index in the UV radiation region at room temperature is about 2.4, the critical angle of total internal reflection is $< 25°$. This fact makes the formation of laser modes highly probable as a result of total internal reflections, as we have mentioned in [1,2] for ZnO rods and tetrapods.

Of certain interest are ZnO structures that represent films composed of several uneven narrowing parts with oblong distorted cones. We refer to such particles as microtowers. In Fig. 1, microphotographs (SEM) of samples 1 and 2 of such films are shown. It should be noted that here the particles have the form of hexagonal prism under the terminal cone. It is very likely that this vary prism may serve as a laser cavity.

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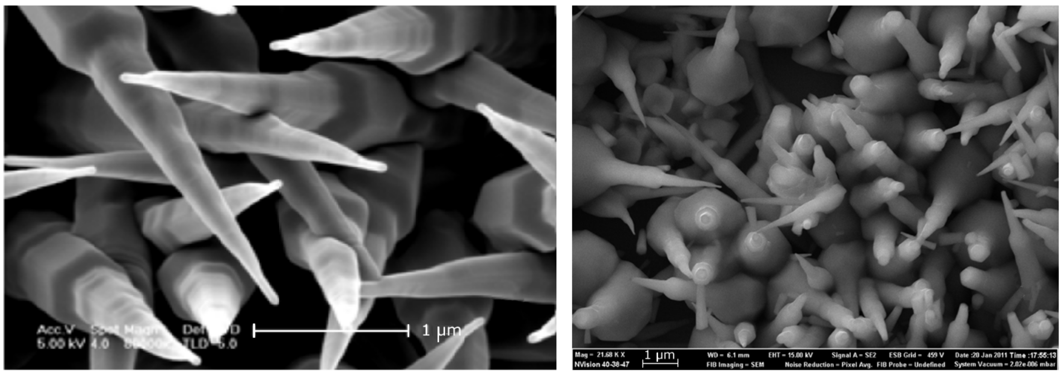
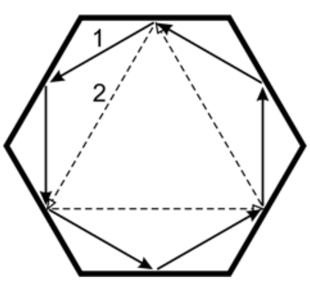


Fig. 1. Microphotographs of microtowers films of sample 1 (left) and sample 2 (right).

One can expect the existence of whispering gallery modes (WGM) in such a laser cavity. The relevant scheme is numbered 1 in Fig. 2 (left) where the mode is formed as a result of six-fold total internal reflections (TIRs). In Fig. 2 (left), a scheme of quasi-WGM is also shown, which is formed as a result of three-fold TIRs with an angle of incidence equal to 30°. There are many scientific publications on WGMs in different ZnO structures, for example, [3–6]. The existence of quasi-WGMs is as demonstrated, for example, in [7]; see Fig. 2 (right) where weak features of the luminescence spectrum are interpreted as a demonstration of quasi-WGMs.



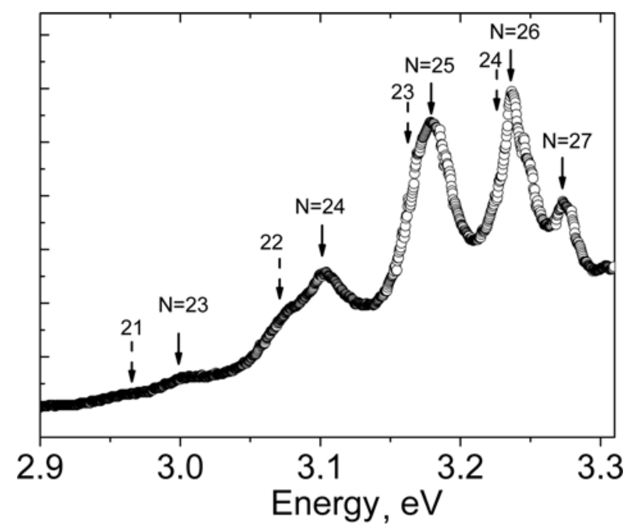


Fig. 2. Schemes of WGM shown by solid arrows 1 and quasi-WGM shown by dashed arrows 2 (left) and the WGMs (solid arrows) and quasi-WGMs (dashed arrows) adopted from [7] (right).

In our experiments, as a rule, two lasing bands with almost constant spectral distance between them are obtained. It is not possible to explain such a result by using only WGMs. In this paper, we will show that one of the two lasing bands in the spectra is due to quasi-WGMs. Thus, from our point of view, this is the first clear demonstration of the existence of quasi-WGMs.

2. Experimental Details

Zinc oxide samples with microtowers were synthesized by the thermal evaporation method in a horizontal tube furnace. The source material of Zn (99.99% purity) powder was loaded into a ceramic boat placed at the end of a one-end sealed quartz tube with 2 cm diameter. Silicon (100) substrates were placed about 40 cm away from the source materials to receive the products. The evaporation of Zn was conducted at 600°C for 60 min under a constant pressure of 76 Pa. Pure argon and oxygen (99.9%) were used as the carrier gas, which was introduced from one end of the quartz tube at a flow rate of 220 and 120 sccm (standard cubic centimeters per minute), respectively. The ZnO microtowers were obtained after the furnace was cooled to room temperature in the natural way.

Laser spectra were measured under pumping by the third harmonics of the Q-switched Nd:YAG laser (355 nm, pulse duration ∼6 ns with repetition rate 15 Hz). The image of the pumping spot was focused on the entrance slit of an MDR-23 monochromator. The spectra were recorded either by a PET-79 or by a Videoscan-285 camera with the CCD matrix (size 8.8×6.6 mm^2) installed in the plane of the exit slit of the monochromator covering the wavelength interval of 11.8 nm with dispersion around 0.0085 nm/pixel. When necessary to obtain a lengthy piece of the spectrum, sequential intervals 11.8 nm were registered with shifts of the scale on 10 nm. After that, pieces measuring 10 nm long were taken from these intervals and sewn together.

3. Experimental Results

Figure 3 shows five spectra obtained with the Videoscan-285 camera. These spectra were registered at different places in sample 1. (All spectra are normalized on shortwave peaks.) It can be clearly seen that the positions of the spectral peaks change slightly from one place to the other, but the spectral distance between the peaks remains practically constant, ∼3 nm. The widths of the bands are approximately 1.5 nm.

In Fig. 4 (left), lengthy pieces of the spectra of sample 1 obtained at the same place under different pumping-energy densities are shown; here it is possible to estimate the lasing threshold, which is between 4.3 and 5 mJ/cm^2. In addition, from this figure it is clear that only a small part of the particles present in the pumping spot is involved in the lasing action. As a result, the main part of the emission in Fig. 4 (left) is luminescence, and the lasing contribution is small.

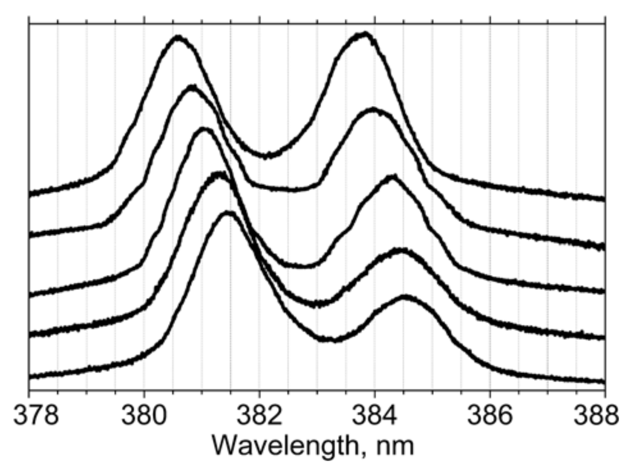


Fig. 3. Laser spectra at different places under fixed pumping level (sample 1).

Different results were obtained for sample 2, its microphotograph is shown in Fig. 1 (right); nevertheless, the results were absolutely the same in terms of quality. Figure 4 (right) shows spectra of this sample obtained under different pumping levels and registered by a PET. One can see that here again there exist two bands. However, they are in another spectral range, and the distance between them is longer, i.e., about 5 nm. The lasing threshold in this sample is about 6 mJ/cm^2; see Fig. 4 (right).

To study the nature of the band widths obtained, the laser spectra were registered under different pumping-spot sizes. In Fig. 5 (left), it is clear that the band widths decrease with decrease in the pumping-spot size. This result is evidence of the width dependence on the number of particles located inside the spot.

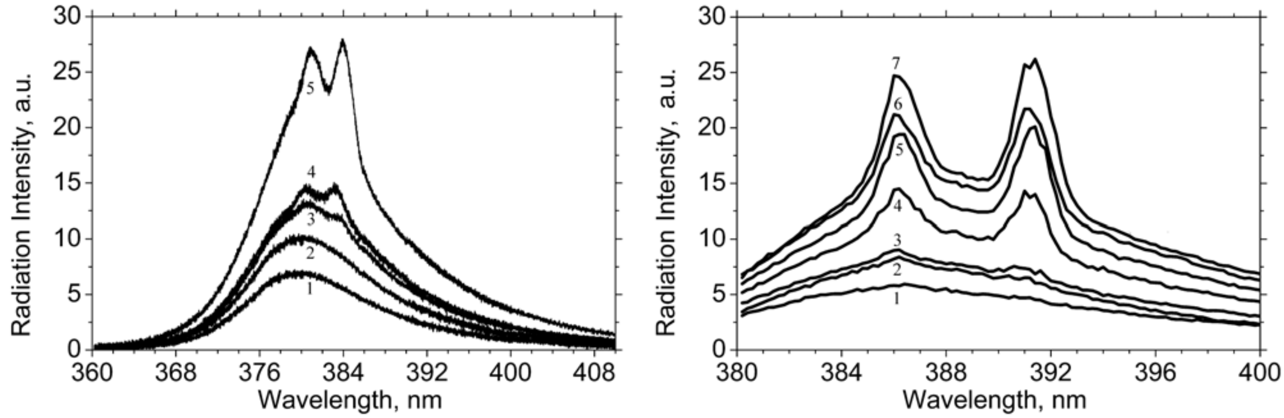


Fig. 4. Spectra of microtowers under different pumping levels 2.6 (1), 4.3 (2), 5.0 (3), 9.0 (4), and 13.1 mJ·cm^{-2} (5) for sample 1 (left) and 4.2 (1), 6.4 (2), 8.1 (3), 11.4 (4), 15.5 (5), 19.1 (6), and 23.7 mJ·cm^{-2} (7) for sample 2 (right).

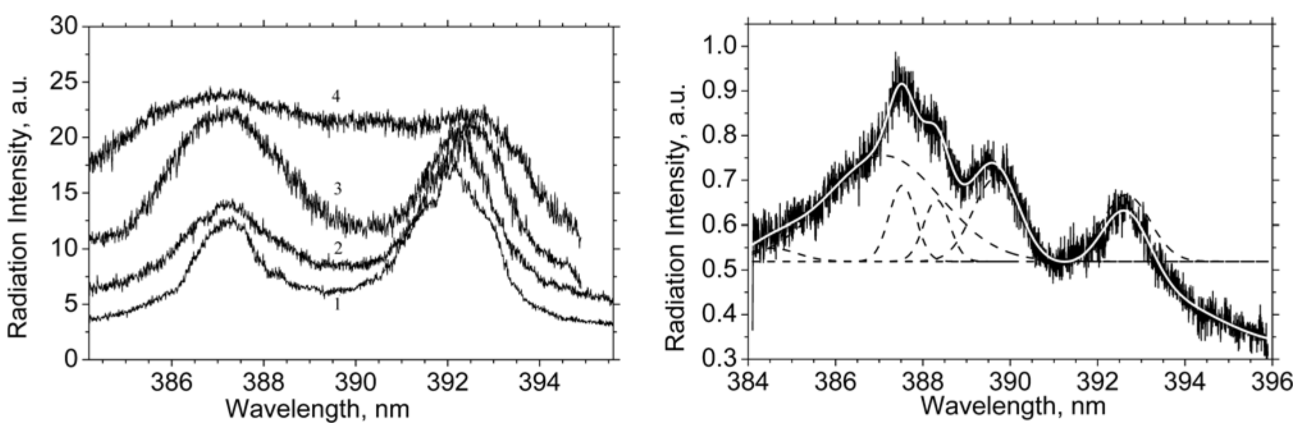


Fig. 5. Laser spectra of sample 2 under different pumping-spot sizes 150 (curve 1), 300 (curve 2), 800 (curve 3), and 1,500 μm (curve 4) (left) and laser spectra under the minimum pumping-spot size with Lorentz components shown by dashed curves (right).

From our point of view, the bands in the spectra are the result of overlapping of lines corresponding to different particles since the particle diameters vary randomly. This conclusion is confirmed by Fig. 5 (right) which shows that when the pumping spot is small enough, the spectral bands are not smooth and can be expanded into Lorentz components. To be sure, a single component does not represent the lasing action of a single microtower.

4. Analysis of the Results

The form of the particles, microtowers, can be considered as composed of three parts. The bottom is a badly shaped cone with a nearly ball-shaped understructure, the middle part is the most perfect and represents a hexagonal prism, and the top of the "tower" is a cone spire (in cross-section, this part has an irregular shape and its lateral surfaces are uneven). It is very likely that the lasing action emerges in the middle part.

Since the cross sections of the most perfect parts of the particles are hexagons, it is natural to suggest that whispering gallery modes (WGMs) participate in the lasing action. As mentioned above, we obtained in the experiment that two bands of lasing action appear together (Figs. 3 and 4); thus, they are probably formed in the same tower. However, the spectral distances between the adjacent WGMs are significantly larger than 3 or 5 nm, hence the suggestion of quasi-WGMs appears, and we will calculate the dependence of the wavelength on the hexagon size for the WGMs and quasi-WGMs. For this purpose, we use the expressions for the wavelength of relevant modes taking into account the formulas for δ phase shift under TIR [8],

$$\tan\frac{\delta_{TE}}{2} = -\frac{\sqrt{\sin^2\theta_i - n^2}}{n^2\cos\theta_i}, \qquad \tan\frac{\delta_{TM}}{2} = -\frac{\sqrt{\sin^2\theta_i - n^2}}{\cos\theta_i},$$

where θ_i is the angle of incidence, TM and TE correspond to the polarization of electric fields parallel and perpendicular to the c axis of ZnO, and n is a parameter inverse to the ZnO refractive index. We will further refer to n as the ZnO refractive index. Based on the resonance condition for the WGM wavelength λ, we write the following expressions:

$$\lambda_{\rm TE} = \frac{3\sqrt{3}\,nR}{N + (6/\pi)\tan^{-1}(n\sqrt{3n^2-4})}, \qquad \lambda_{\rm QTE} = \frac{9nR}{2N + (6/\pi)\tan^{-1}\left(n\sqrt{(n^2-4)/3}\right)},$$

$$\lambda_{\rm TM} = \frac{3\sqrt{3}\,nR}{N + (6/\pi)\tan^{-1}\left(n^{-1}\sqrt{3n^2-4}\right)}, \qquad \lambda_{\rm QTM} = \frac{9nR}{2N + (6/\pi)\tan^{-1}\left(n^{-1}\sqrt{(n^2-4)/3}\right)},$$

where Q stands for quasi-modes, N is the mode number, n is the ZnO refractive index, and R is the side of the hexagonal section or half of its diameter.

The calculated dependences of the wavelength on the diameter for a wide range of modes are shown in Fig. 6; here the quadratic approximation was used for the spectral dependence of the ZnO refractive index [9]. Horizontal lines $P_{1,2}$ indicate the maximum positions of the lasing bands.

From Fig. 6, we can see that the TM8 and QTM8 modes best explain the experimental spectra, and the diameters corresponding to the spectral maxima are in the range of ~590 nm for a spectral distance between the maxima of ~3 nm, and in the range of ~615 nm for a spectral distance of about 5 nm. Based on the microphographs (Fig. 1), one can be sure that, within the margin of error, the diameters of hexagonal parts of samples 1 and 2 are in these ranges. It is noteworthy that even a relatively insignificant change in size changes noticeably the peak positions in the spectrum. Thus, based on the model proposed, one can evaluate the diameter of the hexagonal sections of the microtowers with a rather high degree of accuracy.

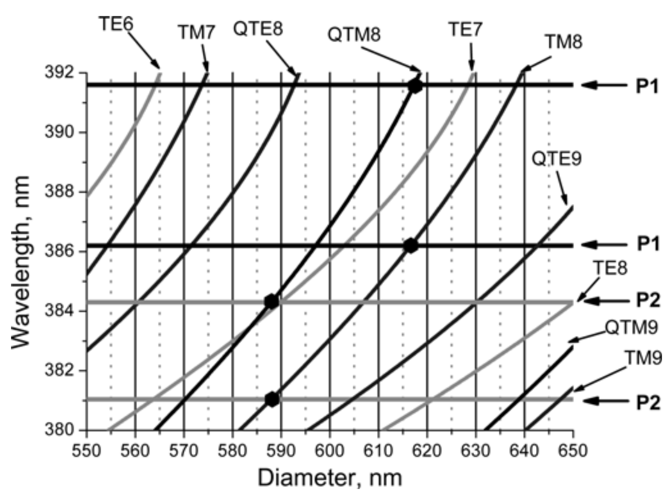


Fig. 6. Dependence of the lasing wavelength on the diameter of the hexagonal section.

The band widths is a result of spreading of the diameters within the pumping spot. This is confirmed by the narrowing of the bands with decrease in the pumping-spot size and by the structuring of the bands under a small pumping spot. The shift of band maximum from one place to the other is also due to the spread of diameters — the position of the diameter-distribution maximum can be different.

5. Conclusions

ZnO films composed of particles that look like microtowers were synthesized on a silicon substrate by a thermal evaporation method using Zn powder as the source material. We have shown that the laser spectra of such films, as a rule, consist of two bands. The positions of the spectral peaks change slightly from one place of the sample to the other, but the spectral distance between the bands remains practically constant, namely, 3 nm for sample 1 and 5 nm for sample 2. Our estimation of the lasing thresholds showed that they are 5 and 6 mJ/cm^2, respectively.

By numerical analysis, we have shown that the short wavelength lasing band is the TM WGM with mode number 8, and the long wavelength lasing band is the TM quasi-WGM with mode number 8. Thus, this is the first clear demonstration of the existence of the quasi-WGMs.

Acknowledgments

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